The effects of disorder in dimerized quantum magnets in mean field approximations

Rakhimov, Abdulla ; Mardonov, Shuhrat ; Ya Sherman, E ; Schilling, Andreas

Abstract: We study theoretically the effects of disorder on Bose–Einstein condensates (BEC) of bosonic triplon quasiparticles in doped dimerized quantum magnets. The condensation occurs in a magnetic field, where the concentration of bosons in the random potential is sufficient to form the condensate. The effect of doping is partly modeled by a -correlated distribution of impurities, which (i) leads to a uniform renormalization of the system parameters and (ii) produces disorder in the system with renormalized parameters. This approach can explain qualitatively the available magnetization data on the Tl1−xKxCuCl3 compound taken as an example. In addition to the magnetization, we found that the speed of the Bogoliubov mode has a maximum as a function of x. No evidence of the pure Bose glass phase has been found in the BEC regime.

DOI: https://doi.org/10.1088/1367-2630/14/11/113010

Originally published at:
Rakhimov, Abdulla; Mardonov, Shuhrat; Ya Sherman, E; Schilling, Andreas (2012). The effects of disorder in dimerized quantum magnets in mean field approximations. New Journal of Physics, 14(11):113010.
DOI: https://doi.org/10.1088/1367-2630/14/11/113010

The effects of disorder in dimerized quantum magnets in mean field approximations

Abdulla Rakhimov[a,b], **Shuhrat Mardonov**[c], **E. Ya. Sherman**[d,e], **Andreas Schilling**[b]

[a] Institute of Nuclear Physics, Tashkent 100214, Uzbekistan
[b] Physik-Institut, University of Zürich, Winterthurerstrasse 190, 8057 Zürich, Switzerland
[c] Samarkand State University, 703004 Samarkand, Uzbekistan
[d] Department of Physical Chemistry, University of Basque Country, 48080 Bilbao, Spain
[e] IKERBASQUE Basque Foundation for Science, 48011 Bibao, Bizkaia, Spain

E-mail: `evgeny_sherman@ehu.es`

Abstract. We study theoretically the effects of disorder on Bose-Einstein condensates (BEC) of bosonic triplon quasiparticles in doped dimerized quantum magnets. The condensation occurs in a strong enough magnetic field, where the concentration of bosons in the random potential is sufficient to form the condensate. The effect of doping is partly modeled by δ - correlated distribution of impurities, which (i) leads to the uniform renormalization of the system parameters and (ii) produces disorder in the system with renormalized parameters. These approaches can explain qualitatively the available magnetization data in the $Tl_{1-x}K_xCuCl_3$ compound taken as an example. In addition to the magnetization, we found that the speed of the Bogoliubov mode has a peak as a function of x. No evidence of the pure Bose glass phase has been found in the BEC regime.

PACS numbers: 75.45.+j, 03.75.Hh

1. Introduction

The effects of disorder on the properties of Bose-Einstein condensates is an interesting problem, both for theoretical and experimental physics [1, 2, 3, 4, 5]. Disorder is important in various systems of real particles such as superfluid ^{4}He, cold atoms in optical lattices, and quasiparticles such as polaritons [6] and excitons [7]. These systems are well-suited for experimental studies, however the theory of disordered ensembles of interacting bosons is complex and there are essentially no exact solutions even in one dimension [8]. To approach this problem, Yukalov and Graham (YG) developed a self-consisted stochastic mean field approximation (MFA) [9] for Bose systems with arbitrary strong interparticle repulsion and arbitrary strength of disorder potential. It was shown that, in general, the Bose system consists of following coexisting components: the condensate fraction, ρ_0, the normal fraction ρ_N, the glassy fraction ρ_G, and, in addition, can be characterized by the superfluid density ρ_s. In the limit of asymptotically weak interactions and disorder the known results, obtained in pioneering work by Huang and Meng [10] (HM) are reproduced by the YG theory. An interesting question here concerns the problem about the existence of a pure Bose glass (BG) phase, i.e. the phase where the condensate fraction is nonzero, while the superfluid fraction is not yet present. Even without disorder, the condensate is depleted by particle-particle interactions and temperature. The inclusion of random fields depletes the condensate further and, possibly, creates the glassy fraction.

As it was understood recently, a new class of BEC can be provided by spin-related quasiparticles in magnetic solids such as intensively pumped magnons [11] or triplons in the dimerized quantum magnets in the equilibrium [12]. In the magnets, the effect of disorder, which can be produced by admixing other chemical elements, can be rather strong to be seen in the properties such as the temperature-dependent magnetization. The so far most investigated compound showing BEC of triplons is $TlCuCl_3$. To study the effect of disorder, solid solutions of antiferromagnetics $TlCuCl_3$ and $KCuCl_3$, i.e. $Tl_{1-x}K_xCuCl_3$ have been experimentally investigated recently [13, 14, 15] at low temperatures T. The zero-field ground states of $TlCuCl_3$ and $KCuCl_3$ are spin singlets with excitation gaps $\Delta_{\rm st} = 7.1$ K and $\Delta_{\rm st} = 31.2$ K, respectively and the magnetic excitations are spin triplets. Triplons arise in magnetic fields $H > H_c$, where H_c is defined by condition of closing the gap by the Zeeman coupling, that is $\Delta_{\rm st} = g\mu_B H_c$, where g is the electron Landé factor and μ_B is the Bohr magneton. In the mixture $Tl_{1-x}K_xCuCl_3$ the magnetization exhibits a cusplike minimum at a critical temperature $T_c(H)$ for fixed magnetic field $H \geq H_c$ similarly to the parent compound, as can be successfully explained in terms of triplon BEC [16, 17, 18].

For a theoretical description it is natural to assume that for weak doping $x \ll 1$ in the mixed system $Tl_{1-x}K_xCuCl_3$ a small admixture of potassium forms a disorder potential. Consequently, the recently developed theories of "dirty bosons" [1, 2, 9, 10] can be applied to study the BEC of triplons in $Tl_{1-x}K_xCuCl_3$. Here the following natural questions arise. For example, what is the correspondence between admixing parameter

x and that of the disorder potential ? What are the experimental consequences of the disorder ? Yamada *et al.* [14] analyzed the electron spin resonance spectrum in $Tl_{1-x}K_xCuCl_3$ and concluded that there is a Bose glass - BEC transition near a critical magnetic field. Although this interpretation might need a further analysis (see discussion in Refs. [19]) it would be interesting to study the influence of the glassy phase, or more exactly, of the glassy fraction ρ_G on the magnetization M. Note that even the existence of a pure Bose glass phase still is a matter of debate even in theoretical approaches. For example, it may be predicted by the approach used by Huang and Meng [10] if one extends their formulas from weak disorder to a strong one. On the other hand, no pure Bose glass was found in Monte - Carlo simulations [20] for atomic gases, but predicted for triplons at $T = 0$ by Nohadani *et al.* [21].

Here we develop a theory of the disorder effects on the BEC of triplons taking $Tl_{1-x}K_xCuCl_3$ as an example for studies of specific properties. For example, in atomic gases considered in Refs. [9, 10], the chemical potential μ is determined self-consistently with fixed number of atoms, while in the triplon gas the chemical potential is a given external parameter controlled by the applied magnetic field and the number of triplons is conserved in the thermodynamic limit. To clarify the terms, we underline that the number of magnons may vary but that of triplons may be tuned and kept fixed, which makes possible the BEC of the latter.

The paper is organized as follows. In Sections II and III we outline the YG and HM approaches valid only for $T \leq T_c$ and extend it for the triplon system. The shift of T_c due to disorder and the normal phase properties will be discussed in Section IV. Our numerical results will be presented in Section V. Conclusions will summarize the results of this work.

2. Yukalov-Graham approximation for disordered triplons

Here we reformulate this approximation to the triplon system with arbitrary disorder. The Hamiltonian energy operator of triplons with contact interaction and implemented disorder potential $V(\mathbf{r})$ is given by

$$H = \int d^3r \left[\psi^\dagger(\mathbf{r}) \left(\hat{K} - \mu + V(\mathbf{r}) \right) \psi(\mathbf{r}) + \frac{U}{2} \left(\psi^\dagger(\mathbf{r})\psi(\mathbf{r}) \right)^2 \right], \tag{1}$$

where $\psi(\mathbf{r})$ is the bosonic field operator, U is the interparticle interaction strength, and $\hat{K}$ is the kinetic energy operator which defines the bare triplon dispersion $\varepsilon_{\mathbf{k}}$. Since the triplon BEC occurs in solids, we perform integration over the unit cell of the crystal with the corresponding momenta defined in the first Brillouin zone. Below the bare spectrum will be assumed as a simple isotropic one: $\varepsilon_{\mathbf{k}} = k^2/2m$, where m is the triplon effective mass. The distribution of random fields is assumed to be zero - centered, $\langle V(\mathbf{r}) \rangle = 0$, and the correlation function $R(\mathbf{r} - \mathbf{r}') = \langle V(\mathbf{r})V(\mathbf{r}') \rangle$. Here and below we adopt the units $k_B \equiv 1$, $\hbar \equiv 1$, and $V \equiv 1$ if not stated otherwise for the unit cell volume.

To describe Bose condensed system where the global gauge symmetry is broken, one employs the Bogoliubov shift:

$$\psi(\mathbf{r}) = \sqrt{\rho_0(\mathbf{r})} + \psi_1(\mathbf{r}) \tag{2}$$

where the condensate density $\rho_0(\mathbf{r})$ is constant for the homogeneous system, $\rho_0(\mathbf{r}) \equiv \rho_0$. Since by the definition the average of $\psi^\dagger(\mathbf{r})\psi(\mathbf{r})$ is the total number of particles:

$$N = \int_V d^3r \langle \psi^\dagger(\mathbf{r})\psi(\mathbf{r})\rangle \tag{3}$$

with the density of triplons per unit cell $\rho = N/V$, from the normalization condition

$$\rho = \rho_0 + \rho_1 \tag{4}$$

one immediately obtains

$$\rho_1 = \frac{1}{V}\int_V d^3r \langle \psi_1^\dagger(\mathbf{r})\psi_1(\mathbf{r})\rangle. \tag{5}$$

Therefore the field operator $\psi_1(\mathbf{r})$ determines the density of uncondensed particles.

The YG approximation is formulated in representative ensemble formalism, which includes two Lagrange multipliers, μ_0 and μ_1, defined as:

$$N_0 = -\frac{\partial \Omega}{\partial \mu_0}, \quad N_1 = -\frac{\partial \Omega}{\partial \mu_1}, \tag{6}$$

where Ω is the grand thermodynamic potential. It was shown that disorder would not change the explicit expressions for chemical potentials, obtained earlier [22] in Hartree-Fock-Bogoliubov (HFB) approximation without disorder,

$$\mu_0 = U(\rho + \rho_1 + \sigma), \quad \mu_1 = U(\rho + \rho_1 - \sigma), \tag{7}$$

where $\sigma = \frac{1}{V}\int_V d^3r \langle \psi_1(\mathbf{r})\psi_1(\mathbf{r})\rangle$ is an anomalous density. The total system chemical potential μ is determined by

$$\mu\rho = \mu_1\rho_1 + \mu_0\rho_0. \tag{8}$$

Clearly, when the gauge symmetry is not broken, i.e. $\rho_0 = 0$, $\sigma = 0$, $\rho_1 = \rho$, both μ_0 and μ_1 coincide giving $\mu = \mu_1 = 2U\rho$.

Now we note again that in contrast to homogeneous atomic gases considered in Refs.[9, 10], where ρ is fixed and $\mu(\rho)$ should be calculated as an output parameter, in the triplon gas the chemical potential is fixed by the external magnetic field, while the density $\rho = \rho(\mu)$ should be calculated self consistently. In fact, in a system of triplons μ characterizes an additional direct contribution to the triplon energy due to the external field H and can be written as

$$\mu = g\mu_B H - \Delta_{\rm st}, \tag{9}$$

which can be interpreted as a chemical potential of the $S_z = -1$ triplons.

The magnetization is proportional to the triplon density

$$M = g\mu_B \rho \tag{10}$$

with ρ is defined from (8) as

$$\rho = \frac{1}{\mu}\left(\mu_1\rho_1 + \mu_0\rho_0\right) \tag{11}$$

where μ_0 and μ_1 are given in (7) and the densities ρ_0, ρ_1 must be calculated self consistently.

It is well known [23] that the disorder field leads to creation of a glassy phase with the density ρ_G. In this approximation each of ρ_1 and σ are presented as

$$\rho_1 = \rho_N + \rho_G; \quad \sigma = \sigma_N + \rho_G \tag{12}$$

where ρ_N and σ_N are the normal and anomalous densities without disorder. In the YG method, based on HFB approximation, the following explicit relations can be obtained [17]:

$$\rho_N = \frac{(\Delta m)^{3/2}}{3\pi^2} + \int \frac{d^3k}{(2\pi)^3} f_B(\mathcal{E}_k)\frac{\varepsilon_k + \Delta}{\mathcal{E}_k}, \tag{13}$$

$$\sigma_N = \frac{(\Delta m)^{3/2}}{\pi^2} - \Delta \int \frac{d^3k}{(2\pi)^3} f_B(\mathcal{E}_k)\frac{1}{\mathcal{E}_k}, \tag{14}$$

with the Bose distribution of Bogoliubov excitations $f_B(\mathcal{E}_k) = 1/(e^{\mathcal{E}_k/T} - 1)$ having the dispersion $\mathcal{E}_k$

$$\mathcal{E}_k = \sqrt{\varepsilon_k}\sqrt{\varepsilon_k + 2\Delta}. \tag{15}$$

For small momentum k the dispersion is linear, $\mathcal{E}_k = ck$, and the speed of the Bogoliubov mode

$$c = \frac{\sqrt{\Delta}}{\sqrt{m}}. \tag{16}$$

The self energy Δ is determined formally by the same equation as in the case when the disorder is neglected,

$$\Delta = U(\rho_0 + \sigma) = U(\rho - \rho_N + \sigma_N). \tag{17}$$

The contribution from the disorder potential is hidden in the density of the glassy fraction

$$\rho_G = \frac{1}{V}\int_V d^3r \langle\langle \psi_1(\mathbf{r})\psi_1(\mathbf{r})\rangle\rangle \tag{18}$$

where the double angle brackets mean stochastic average in the sense of Ref.[9]. In general, the calculation of ρ_G is rather complicated, but for the δ - correlated disorder i.e. for the white noise,

$$\langle\langle V(\mathbf{r})V(\mathbf{r}')\rangle\rangle = R\delta(\mathbf{r} - \mathbf{r}'), \tag{19}$$

equation (18) may be simplified as [9]

$$\rho_G = \frac{R_0(\rho - \rho_N)}{R_0 + 7(1 - R_0)^{3/7}}. \tag{20}$$

The density of condensed fraction can be found by inserting (20) into the normalization condition (4). The result is

$$\rho_0 = \frac{7(1-R_0)^{3/7}(\rho-\rho_N)}{R_0+7(1-R_0)^{3/7}}. \tag{21}$$

In Eqs. (20) and (21) we introduced the dimensionless parameter R_0 as

$$R_0 \equiv \frac{7Rm^2}{4\pi\sqrt{m\Delta}}. \tag{22}$$

One can see from Eqs. (20) and (21) that the glassy fraction is proportional to the condensed one,

$$\rho_G = \frac{\rho_0 R_0}{7(1-R_0)^{3/7}}. \tag{23}$$

The system of Eqs. (7), (8), (13)-(19) are the basic equations of YG approximation.

Note that YG approach is valid for arbitrary strength of the interaction potential U, and for arbitrary strong disorder. For the weak interactions it leads to pioneering Huang-Meng approach [10], which will be extended to the "dirty triplons" in the next section.

3. Huang-Meng approximation

For completeness, we present here the results for the Huang-Meng approach, based on the so called Hartree Fock Popov (HFP) approximation which has been widely applied in the literature to describe the BEC of triplons [16, 18]. The basic equations of this approach can be obtained by neglecting the anomalous density σ, which leads naturally to the single chemical potential $\mu = \mu_0 = \mu_1$. Namely, one finds from (7), (8) and (17)

$$\Delta = U\rho_0, \qquad \mu = U(\rho+\rho_1). \tag{24}$$

From these equations and (12) one obtains following main equations for the self energy Δ:

$$\Delta = \mu - 2U(\rho_N+\rho_G), \tag{25}$$

where ρ_N is formally given in (13), and ρ_0 is determined by the first equation in (24). The glassy fraction can be obtained from (20) in the linear approximation by R assuming weakness of interparticle interaction [9, 10]

$$\rho_G = \frac{m^2 R}{8\pi^{3/2}}\left(\frac{\rho_0}{a_s}\right)^{1/2}, \tag{26}$$

where $a_s = Um/4\pi$ is the s - wave scattering length. Inserting (26) into (25) we can rewrite the former as

$$\Delta = \mu - 2U\rho_N - \frac{m^2 R\sqrt{\Delta}}{2\pi\sqrt{m}}. \tag{27}$$

To evaluate the densities one has to solve nonlinear algebraic equation (27), where ρ_N is given formally by (13), with respect to Δ. Next, by inserting the result into (24) and (26) one obtains the density of condensed triplons ρ_0 and the glassy fraction ρ_G, respectively. The total density can be evaluated then by the normalization condition $\rho = \rho_0 + \rho_N + \rho_G$.

Another interesting quantity, crucial for determining the Bose glass phase, is the superfluid density, ρ_s. In general it is defined as a partial density appearing as a response to a velocity boost

$$\rho_s = \frac{1}{3mV} \lim_{\mathbf{v}\to 0} \frac{\partial}{\partial \mathbf{v}} \langle \hat{P}_{\mathbf{v}} \rangle \tag{28}$$

where $\hat{\mathbf{P}}_{\mathbf{v}}$ is the total momentum of the system, dependent on the macroscopic velocity $\mathbf{v}$. Referring the reader to original papers [9, 10] we bring below analytical expression obtained there for ρ_s in the case of white noise random potential

$$\rho_s = \rho - \frac{4\rho_G}{3} - \frac{2Q_N}{3T} \tag{29}$$

$$Q_N = \frac{1}{8m} \int \frac{k^2 d^3k}{(2\pi)^3 \sinh^2(\mathcal{E}_k/2T)}, \tag{30}$$

which are formally the same in both approximations.

4. The shift of the critical temperature due to disorder and the $T > T_c$ regime

It is well known that the critical temperature of BEC, T_c for an ideal gas is given by:

$$T_c^0 = \frac{2\pi}{m} \left(\frac{\rho_c}{\zeta(3/2)} \right)^{2/3}, \tag{31}$$

where ρ_c is the total density of triplons near the critical temperature of BEC for pure system,

$$\rho_c = \mu/2U, \tag{32}$$

with $\zeta(x)$ being the Riemann function. Eq. (32) is directly follows, e.g. from Eqs. (7), (8) or (25) by setting $\rho_N = \rho$ and $\rho_0 = \rho_G = 0$.

Clearly, any type of interaction is expected to modify T_c. In general, these modifications are related to the interparticle interactions as well as to the disorder potential. Both approaches, considered here give a zero shift due to the boson-boson repulsion. However the shift due to the $\delta-$correlated disorder (19), $\Delta T_c = T_c - T_c^0$ is given as [9, 24]

$$\frac{\Delta T_c}{T_c^0} = -\frac{2\nu}{9\pi}, \tag{33}$$

where the dimensionless disorder parameter ν

$$\nu \equiv \frac{1}{\rho_c^{1/3} L_{\text{loc}}} \tag{34}$$

is introduced with the localization length

$$L_{\text{loc}} = \frac{4\pi}{7m^2 R}. \tag{35}$$

For practical calculations we rewrite T_c in Eq. (33), which is in a good agreement with perturbative estimates [2] as well as with Monte Carlo simulations [25], as an explicit function of effective mass m, the interaction strength U , critical magnetic field H_c, disorder parameter ν, and external field H as follows:

$$T_c = \frac{9\pi - 2\nu}{9m} \left(\frac{\sqrt{2} g \mu_B (H - H_c)}{U \zeta(3/2)} \right)^{2/3} . \tag{36}$$

Now we pass to consider the triplon density in the normal state in the $T - T_c \gg \Delta T_c$ temperature range. The dirty bosons in the normal phase where the gauge symmetry is not broken, are yet poorly studied. For $R = 0$ with $\rho_0 = \rho_G = \sigma = 0$ the triplon gas behaves like an "ideal gas" with an effective chemical potential μ_{eff}, and the density [26]

$$\rho(T > T_c) = \int \frac{d^3k}{(2\pi)^3} \frac{1}{\exp\left((\varepsilon_k - \mu_{\text{eff}})/T\right) - 1}. \tag{37}$$

Although, μ_{eff} is not fairly known it depends in general, on ρ, as well as on R. For the pure case MFA [26] gives $\mu_{\text{eff}}(R = 0) = \mu - 2U\rho$. The contribution from the disorder potential has been studied neither in YM nor in HM approaches. Therefore, to make the calculations self consistently, we have to use

$$\rho(T > T_c) = \int \frac{d^3k}{(2\pi)^3} \frac{1}{\exp\left((\varepsilon_k - \mu - 2U\rho)/T\right) - 1}, \tag{38}$$

which yields the density ρ as a solution of the nonlinear equation (38).

5. Results and discussions

In the calculations below, the energies are measured in Kelvin, the mass in K^{-1}, the densities are dimensionless and the Bohr magneton is $\mu_B = 0.671668$ K/T. As to the strength of disorder potential R, it has units K^{-2} while the disorder parameter ν, defined in Eq. (34) is a number supposed to be less than one, $\nu < 1$. As a material parameter, we use mean dimer-dimer distance in $TlCuCl_3$ $r_{dd} = 0.79$ nm [18].

To perform numerical calculations in the YG approximation, assuming that μ, U, m, and R are given parameters, we used following strategy. (i) By inserting (7), (12), (20) and (21) into (11) one gets quadratic algebraic equation with respect to ρ and solves it analytically. (ii) By using this $\rho(\mu, R, \Delta)$ and (13), (14) in (17) we solve the latter numerically with respect to Δ, and (iii) By insert back this Δ into $\rho(\mu, R, \Delta)$ to find the magnetization from (10) and evaluate other densities like ρ_0 and ρ_G from Eqs. (20), (21).

In Figure 1 we present as an example the total triplon density $\rho(T)$ for a clean and strongly disordered ($\nu = 0.45$, see Eq.(34)) $TlCuCl_3$, obtained in the YG approximation assuming that the total effect of the doping leads only to the randomness in the triplon subsystem.

The calculation of other quantities using the same assumption shows that the disorder leads to a decrease in the condensed and superfluid fractions, thereby increasing

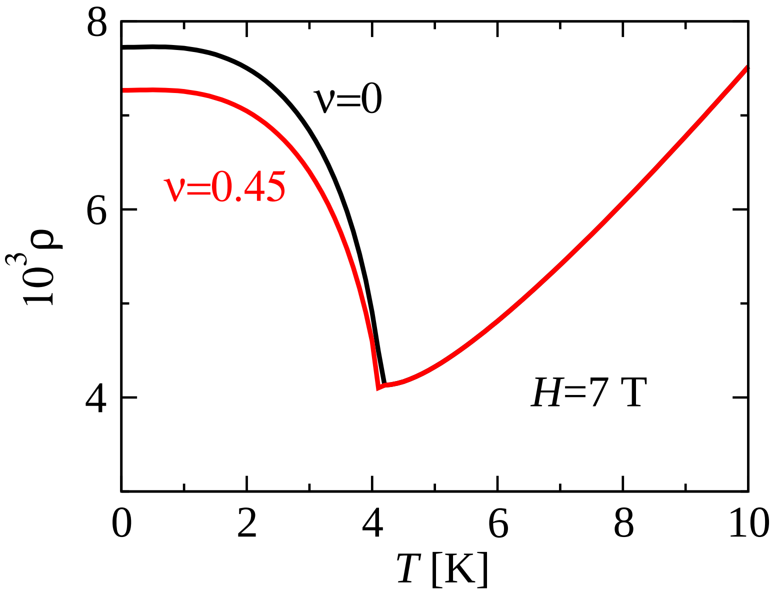

Figure 1. The total triplon density as a function of temperature in the YG approximation for two values of ν. Here following set of parameters m=0.0204 K^{-1}, $\Delta_{\rm st} = 7.3$ K, $U = 313$ K, and $g = 2.06$ [16] valid for pure $TlCuCl_3$ is used.

the glassy one. This tendency is quite natural, since the localization effects prevent particles from going into BEC. However, the increase in ρ_G is so weak that along with ρ_0 the total number of triplons ρ is also decreased with increasing the strength of disorder potential R. Bearing in mind that ρ is proportional to the magnetization M, and ν is assumed to be approximately proportional to x, and comparing Fig.1 with the experimental data illustrated in Fig.2 one may conclude that the agreement between the theory and the experiment is unsatisfactory since the main features of the experimental results are not reproduced there. As it is seen in Fig.2 the disorder leads to an increase in the magnetization and, hence, in the total triplon density. This is accompanied by the decrease in the transition temperature. We therefore conclude that while the triplon gas can be considered similarly as atomic gases for which the considered mean-field approximations were developed, some further additional specific material - related properties of the dirty boson problem in quantum magnets must be taken into account.

First we note that the singlet - triplet excitation gap $\Delta_{\rm st}$, proportional to the critical field, H_c, decreases under high pressure. This was experimentally observed in Ref.[27] for the pure spin system $TlCuCl_3$. On the other hand it can be argued that the doping acts as a chemical pressure, which decreases H_c. In fact, since the ionic radius of K^+ is smaller than that of Tl^+, a partial substitution of Tl^+ ions with K^+ ions produces not only the exchange randomness, but also a compression of the crystal lattice. Thus the increase of the doping parameter, x, leads to decrease in H_c which has indeed been observed experimentally [13, 28, 29]. Second, the disorder may increase the triplon effective mass thereby decreasing the critical temperature T_c even when the gap decreases (similar effects were observed for helium in porous media [30, 31]). Note that this effect manifests

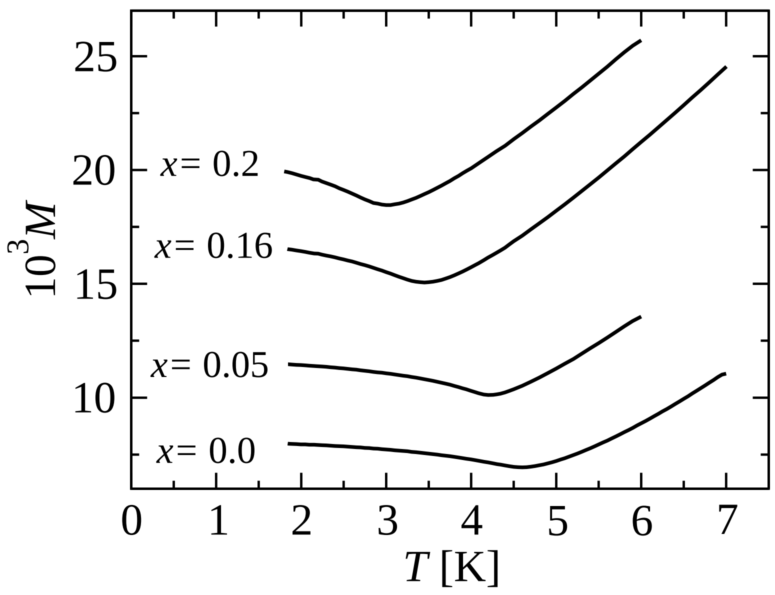

Figure 2. The experimental low temperature magnetization in units of Bohr magneton per Cu ion of $Tl_{1-x}K_xCuCl_3$ for various x in $H = 7$ T magnetic field. (Reproduced from Ref.[13]).

itself in different ways. For example, for the mixed compound IPACu$(Cl_xBr_{1-x})_3$ the critical field, H_c remains almost unchanged with varying x and then, abruptly becomes zero near the Cl-rich phase [32]. In another triplon-BEC compound, $Ni(Cl_{1-x}Br_x)_2$-$4SC(NH_2)_2$, it decreases by a factor of two when x changes from zero to 0.08 [29] although the physics of this decrease can be different from that in $Tl_{1-x}K_xCuCl_3$ due to the fact that Br atomic radius is larger than the atomic radius of Cl. These effects of renormalization of the triplon spectrum by disorder can be considered similarly to the virtual crystal approximation in the simulations of disorder in solids, where the disorder is assumed to lead to a uniform change in the system parameters. The effects of disorder such as the appearance of the glassy phase with the density ρ_G and related phenomena manifest itself in addition to these uniform changes.

The phase diagram of $Tl_{1-x}K_xCuCl_3$ in the (H, T) plane was experimentally determined in Refs.[13, 28] for various doping x, and the critical field H_c was also estimated by extrapolation to zero temperature. In the present work the $T_c(H)$ dependence is given by Eq. (36). We made an attempt to least - square fit our parameters m and ν by using Eq. (36) to describe the experimental phase diagram. For simplicity we assume that interparticle interaction is not changed by doping, i.e. $U = U(R = 0) = 313$ K. The parameters obtained by this optimization are presented in Table 1.

Having fixed the input parameters for certain values of x, we are now in the position of recalculating the densities as well the magnetization to compare them with the experiment. Figure 3 shows that the doping decreases ρ_0 and ρ_s, and increases ρ_G as it is expected due to the introduced disorder. Due to change of H_c with x, the total

Table 1. Optimized values of the input parameters of the model: the critical field, H_c (taken from Ref. [13]), the disorder parameter ν, and the effective mass m for various doping x. The critical density, ρ_c, the healing length, $\lambda = 1/\sqrt{2m\mu}$, the interparticle distance, $d = 1/\rho_c^{1/3}$ and the localization length, $L_{\rm loc} = d/\nu$ are estimated at $H = 7$ T. It is assumed that the doping effects does not modify the Landé factor g and U.

x	ν	H_c [T]	m [1/K]	U [K]	$\Delta_{\rm st}$ [K]	ρ_c	λ [nm]	d [nm]	$L_{\rm loc}$ [nm]
0	0	5.3	0.0204	313	7.3	0.00376	2.548	5.079	∞
0.05	0.163	4.8	0.0242	313	6.6	0.00493	2.044	4.642	28.364
0.08	0.246	4.4	0.0291	313	6.08	0.00589	1.705	4.375	17.727
0.16	0.476	4.1	0.0392	313	5.6	0.00652	1.396	4.227	8.864
0.2	0.586	3.9	0.0442	313	5.4	0.00684	1.283	4.161	7.091

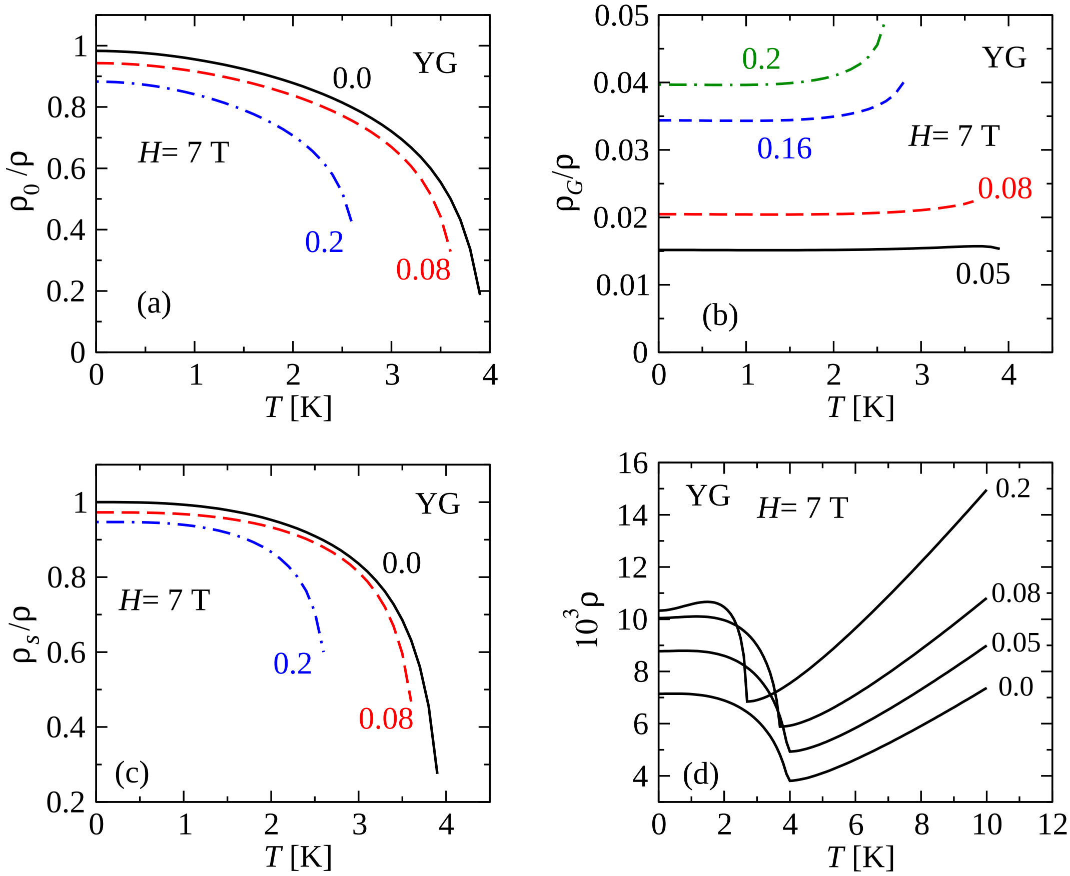

Figure 3. The condensed (a), glassy (b), and superfluid fractions (c) as functions of temperature in the YG approximation for various x marked near the plots with the input parameters from Table 1. The total density of triplons is shown in Fig.3(d).

density of triplons and hence the magnetization, now increases with increasing x in accordance with the experiment. One may conclude that the YG approach may well describe the effect of disorder to the magnetization, with the additional assumption of an x dependence of the effective mass and the critical field.

One of the main characteristics of Bose condensed systems is the speed of the

Bogoliubov mode c, defined here by the Eq. (16), which characterizes the propagation of collective excitations in the condensate. It is interesting to mention that the magnitude of c is large, being only an order of magnitude less than the speed of sound in the crystal. This is due to very small triplon mass in $TlCuCl_3$. Clearly, disorder modifies the small-momentum excitation spectrum of the BEC. Estimates of such modification, $\Delta c = c - c_0$, where c_0 is the speed of the Bogoliubov mode for the pure system, that exist in the literature are controversial. For example, perturbative [1] and hydrodynamic [3] approaches give $\Delta c > 0$, while $\Delta c < 0$ was predicted in Refs. [4, 5]. In Fig. 4 we present the corresponding speed for various doping parameters. It can be seen from comparison of Fig.4(a) and Fig.4(b) that both MFA approximations considered here show the decrease in c with increasing the disorder strength due to the localization effects. However, the effect of disorder is small leading to a less than 10 percent decrease in the speed of the Bogoliubov mode.

However, when the spectrum modification by disorder is also taken into account by a renormalization of the triplon mass and the gap, as close to the real situation, the dispersion of the sound-like mode in fixed magnetic field slightly increases with increasing disorder, reaches a maximum and then starts to decrease, (see Fig.4b). This behavior is caused by interplay between renormalization of the system parameters and localization effects. The former tends to increase c, e.g. by increasing μ, and therefore increasing the density, while the latter tends to decrease c, e.g. by decreasing the condensed fraction. Note that an increase in c with increasing the density was experimentally observed by Andrews *et al.* [33] for the BEC of sodium atoms. This interplay is illustrated in Fig.4(c) for ρ_0 and ρ_s. It can be seen that uniform spectrum renormalization first leads to an antidepletion effect, increasing these quantities, while the localization effects impair the condensation and superfluidity.

We now consider the question about the existence of a pure Bose glass phase at $T = 0$, which, strictly speaking, should fulfill the following criteria [9, 23, 34]: (i) gapless in the excitation spectrum, (ii) insulating behavior, i.e. the superfluid fraction, $\rho_s = 0$, (iii) finite compressibility, and (iv) finite density of states.

In 1970, Tachiki and Yamada [35] have shown that the Heisenberg-like Hamiltonian of $s = 1/2$ dimers can be rewritten as an effective Bosonic Hamiltonian. Recently, Roscilde and Haas [36] generalized this bosonization procedure taking into account disorder and derived a Bose - Hubbard like Hamiltonian usually applied to study "dirty bosons" in optical lattices. So, applying Fishers ideas [23] we may expect a pure Bose glass phase for doped magnets e.g. for $Tl_{1-x}K_xCuCl_3$. Although Monte Carlo calculations [21, 36] confirmed its existence the experimental confirmation is still a matter of debate [19, 37] . We underline here that these Bose glass phases are localized out of the BEC phase, i.e. for $H < H_c$. However, in the present work we have been mainly concentrating on the region with $H \geq H_c$ where the gapless phase can be realized only within the BEC phase. For this case the definition of BG phase may be simplified as a phase with $\rho_0 \neq 0$ and $\rho_s = 0$ since the spectrum of the BEC is gapless by itself. In searching for a such phase we studied ρ_s and ρ_0 at $T = 0$ for various $H \geq H_c$ and x

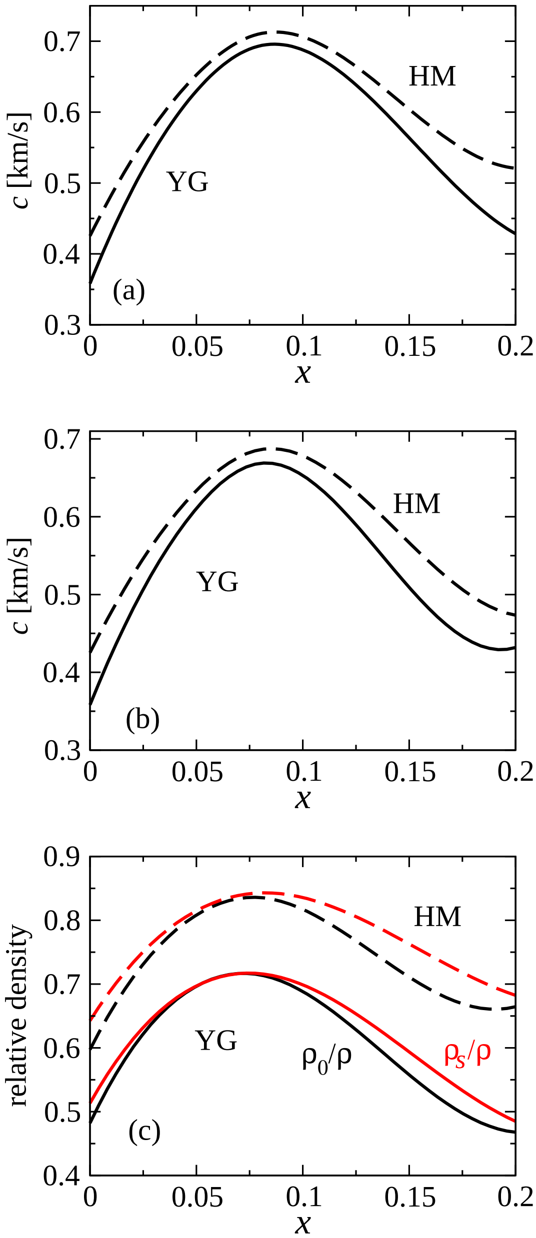

Figure 4. (a) The speed of sound-like condensate mode at $T = 2$ K and $H = 6$ T as a function of doping parameter x, taking into account solely renormalization of the system parameters in Table 1 in the YG (solid line) and the HM (dashed line) approaches. (b) The same as in Fig. 4(a), now with effects of disorder taken into account. (c) The superfluid and condensed fractions (as marked near the plots) in the YG (solid lines) and the HM (dashed lines) approximations with the renormalized bare spectrum parameters.

and found no pure BG phase with $\rho_s = 0$ as illustrated in Fig. 5.

Note also that, as it is seen from Table 1, for moderate values of x considered here, the localization length, i.e the mean free path [38] is larger than interparticle distance, $L_{\rm loc} > d$. Studying the BG phase for $H < H_c$ cases will be the subject of a separate paper.

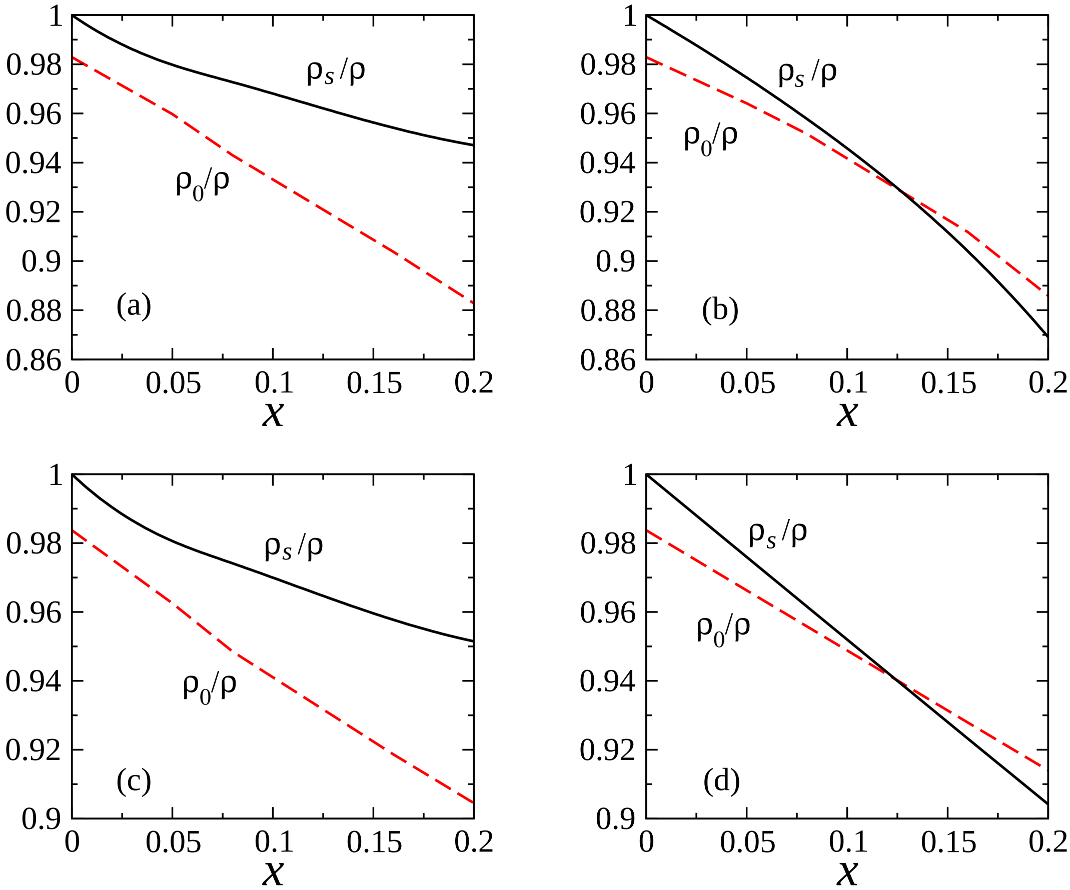

Figure 5. The superfluid, ρ_s/ρ (solid lines) and condensed, ρ_0/ρ (dashed lines), fractions as a function of the doping parameter x at $T = 0$, $H = 7$ T. Upper panel corresponds to the YG approximation and lower panel corresponds to the HM approximation presented here for comparison. Graphs in plots (a), (c) were calculated without bare spectrum renormalization, while graphs in plots (b), (d) take into account spectrum renormalization presented in Table 1.

6. Conclusions

In conclusion, we reformulated and applied two existing MFA approximations for the "dirty boson" problem, to study properties of $Tl_{1-x}K_xCuCl_3$. We showed that these approximations can explain the magnetization data qualitatively with a certain modification of the parameters used in the model similar to the virtual crystal approximation taken into account.

In fact, we have shown that bond random effects in mixed magnetic compounds manifest themselves in dual way: (i) by modification of internal parameters and (ii) by localization on random scatterers. Each of these effects could be studied separately in an appropriate theory, but they should be taken into account simultaneously for an adequate description of the measured magnetization data. The interplay between these effects leads to a nontrivial behavior of the sound-like speed: when H is fixed but x is experimentally varied, it increases for small x, reaches a maximum value and then decreases due to localization effects. While the speed of this mode was measured in dilute BEC of sodium atoms a long time ago [33], it has never been an intense focus of research in dimerized quantum magnets [39]. It could be systematically studied, for example, by measuring the dispersion relation of the Bogoluibov mode with inelastic neutron scattering techniques.

Acknowledgments

The work is partly supported by the Schweizerische Nationalfonds zur Förderung der wissenschaftlichen Forschung under Grants No. IZK0Z2-139441 and No.20-140465. The work of EYS was supported by the MCINN of Spain grant FIS 2009-12773-C02-01, "Grupos Consolidados UPV/EHU del Gobierno Vasco" grant IT-472-10, and by the UPV/EHU under program UFI 11/55. We are indebted to H. Tanaka for providing us with tabulated data for Fig.2.

[1] Falco G M, Pelster A and Graham R 2007 *Phys. Rev.* A **75** 063619
[2] Lopatin A V and Vinokur V M 2002 *Phys. Rev. Lett.* **88** 235503
[3] Giorgini S, Pitaevskii L and Stringari S 1994 *Phys. Rev.* B **49** 12938
[4] Gaul C, Renner N and Müller C A 2009 *Phys. Rev.* A **80** 053620; Gaul C and Müller C A 2011 *Phys. Rev.* A **83** 063629
[5] Zhang L 1993 *Phys. Rev.* B **47** 14364.
[6] Shelykh I A, Kavokin A V, Rubo Y G, Liew T C H and Malpuech G 2010 *Semicond. Sci. Technol.* **25** 013001 and references therein.
[7] Butov L V, Gossard A C and Chemla D S 2002 *Nature* **418** 751
[8] See recent topical review: Shapiro B 2012 J. Phys. A **45** 143001
[9] Yukalov V I and Graham R *Phys. Rev.* A **75** 023619 2007 ; Yukalov V I, Yukalova E P, Krutitsky K V and Graham R 2007 *Phys. Rev.* A **76** 053623
[10] K. Huang and H. F. Meng 1992 *Phys. Rev. Lett.* **69** 644
[11] Demokritov S O, Demidov V E, Dzyapko O, Melkov G A, Serga A A, Hillebrands B and Slavin A N 2006 *Nature* **443** 430
[12] Giamarchi T, Rüegg C and Tchernyshyov O 2008 *Nature Phys.* **4** 198
[13] Oosawa A and Tanaka H 2002 *Phys. Rev.* B **65** 184437
[14] Yamada F, Tanaka H, Ono T and Nojiri H 2011 *Phys. Rev.* B **83** 020409
[15] Tanaka H, Shindo Y and Oosawa A 2005 *Progress of Theoretical Physics Supplement* 159, 189
[16] Yamada F, Ono T, Tanaka H, Misguich G, Oshikawa M and Sakakibara T 2008 *J. Phys. Soc. Jpn.* **77** 013701
[17] Rakhimov A, Mardonov S and Sherman E Ya 2011 *Ann. Phys.* **326** 2499; Rakhimov A, Sherman E Ya and Kim C K 2010 *Phys. Rev.* B **81** 020407
[18] Dell'Amore R, Schilling A, and Krämer K 2009 *Phys. Rev.* B **79** 014438; Dell'Amore R, Schilling A and Krämer K 2008 *Phys. Rev.* B **78** 224403
[19] Zheludev A and Hüvonen D 2011 *Phys. Rev.* B **83** 216401; Yamada F, Tanaka H, Ono T and Nojiri H 2011 *Phys. Rev.* B **83** 216402
[20] Astrakharchik G E, Boronat J, Casulleras J and Giorgini S 2002 *Phys. Rev.* A **66** 023603
[21] Nohadani O, Wessel S and Haas S 2005 *Phys. Rev. Lett.* **95** 227201
[22] Yukalov V I 2008 *Ann. Phys.* **323** 461
[23] Fisher M P A, Weichman P B, Grinstein G and Fisher D S 1989 *Phys. Rev.* B **40** 546
[24] Yukalov V I 2011 *Physics of Particles and Nuclei* **42** 460
[25] Pilati S, Giorgini S and Prokofev N 2009 *Phys. Rev. Lett.* **102** 150402
[26] Nikuni T, Oshikawa M, Oosawa A and Tanaka H 2000 *Phys. Rev. Lett.* **84** 5868

[27] Tanaka H, Goto K, Fujisawa M, Oho T and Uwatoko Y 2003 *Physica* B **329-333** 697; Goto K, Fujisawa M, Tanaka H, Uwatoko Y, Oosawa A, Osakabe T and Kakurai K 2006 *J. Phys. Soc. Jpn.* **75** 064703; Goto K, Osakabe T, Kakurai K, Uwatoko Y, Oosawa A, Kawakami J and Tanaka H 2007 *J. Phys. Soc. Jpn.* **76** 053704
[28] Shindo Y and Tanaka H 2004 *J. Phys. Soc. Jpn.* **73** 2642
[29] Yu R *et al.* 2011 arXiv:1109.4403; Paduan-Filho A 2012 arXiv:1206.0035
[30] Chan M H W, Blum K I, Murphy S Q, Wong G K S and Reppy J D 1988 *Phys. Rev. Lett.* **61** 1950
[31] Shibayama Y and Shirahama K 2011 *J. Phys. Soc. Jpn.* **80** 084604
[32] Manaka H, Yamada I, Mitamura H and Goto T 2002 *Phys. Rev.* B **66** 064402
[33] Andrews M R, Kurn D M, Miesner H-J, Durfee D S, Townsend C G, Inouye S and Ketterle W 1997 *Phys. Rev. Lett.* **79** 553
[34] Yukalov V I 2009 *Laser Physics* **19** 1
[35] Tachiki M and Yamada T 1970 *J. Phys. Soc. Jpn.* **28** 1413
[36] Roscilde T and Haas S. 2006 J. Phys. B **39** S 153
[37] Hong T, Zheludev A, Manaka H and Regnault L-P 2010 *Phys. Rev.* B **81** 060410
[38] Falco G M, Nattermann T and Pokrovsky V L 2009 *Phys. Rev.* B **80** 104515
[39] Schilling A, Grundman H and Dell'Amore R 2011 arXiv:1107.4335